Electronic Supplementary Information

Bifunctional catalysts of Ni nanoparticles coupled MoO_2 nanorods for overall water splitting

Zhiqiang Yao[1,2], Chenfeng Wang[1,2], Zengyao Wang[1,2], Guanglei Liu[1,3], Crystal Bowers[4], Pei Dong[4], Mingxin Ye[1], Jianfeng Shen[1*]*

[1]Institute of Special Materials and Technology, Fudan University, Shanghai 200433, China

[2]Department of Chemistry, Fudan University, Shanghai, 200433, P. R. China

[3]Department of Materials Science, Fudan University, Shanghai, 200433, P. R. China

[4]Department of Mechanical Engineering, George Mason University, VA 22030, USA

[*]Correspondence to: mxye@fudan.edu.cn, jfshen@fudan.edu.cn

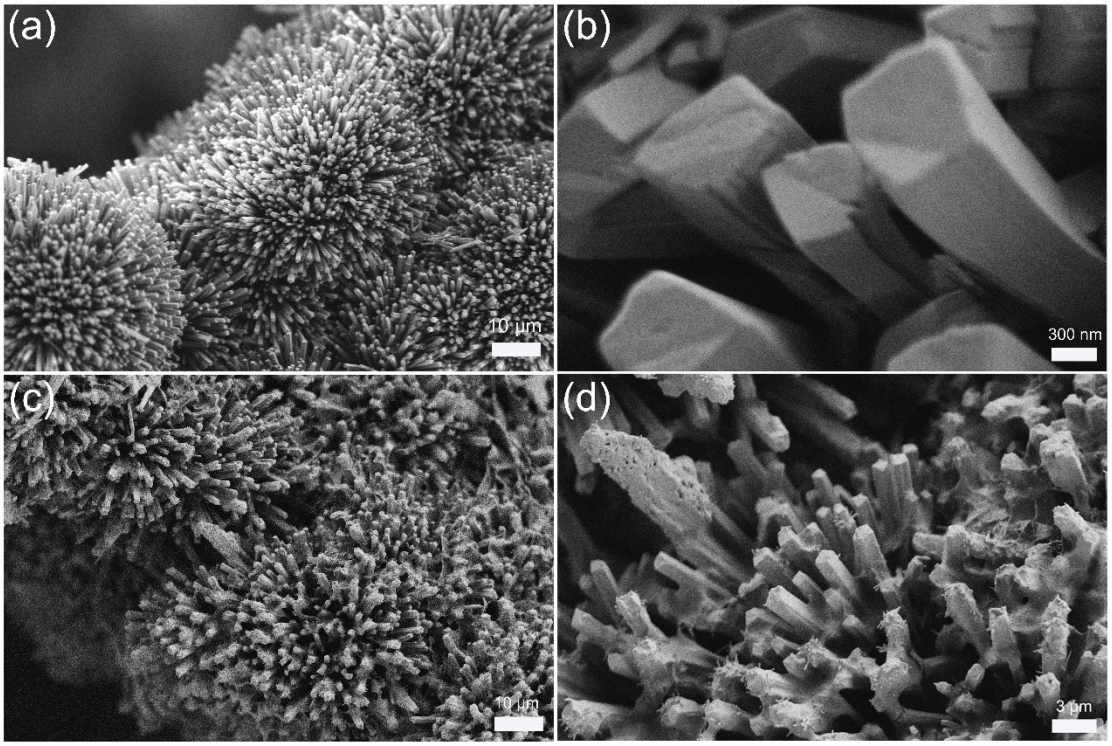

Fig. S1. SEM images of (a, b) $NiMoO_4 \cdot xH_2O$ and (c, d) NMO-ZIF.

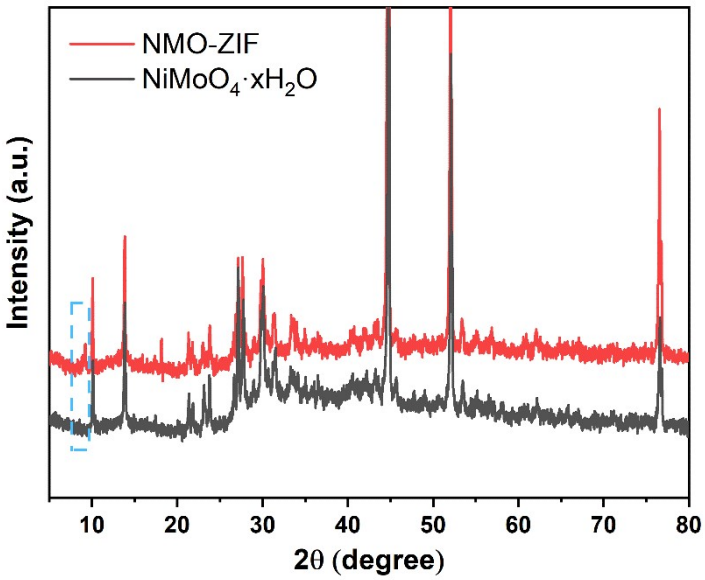

Fig. S2. XRD images of $NiMoO_4 \cdot xH_2O$ and NMO-ZIF.

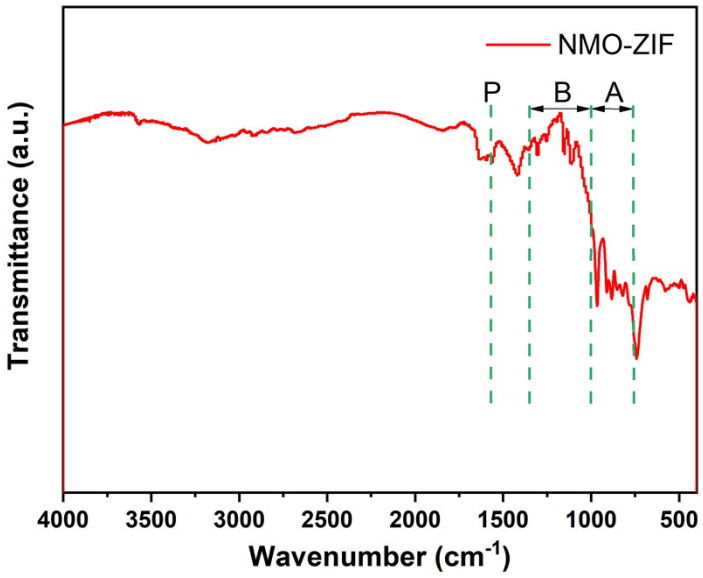

Fig. S3. FTIR spectrum of NMO-ZIF.

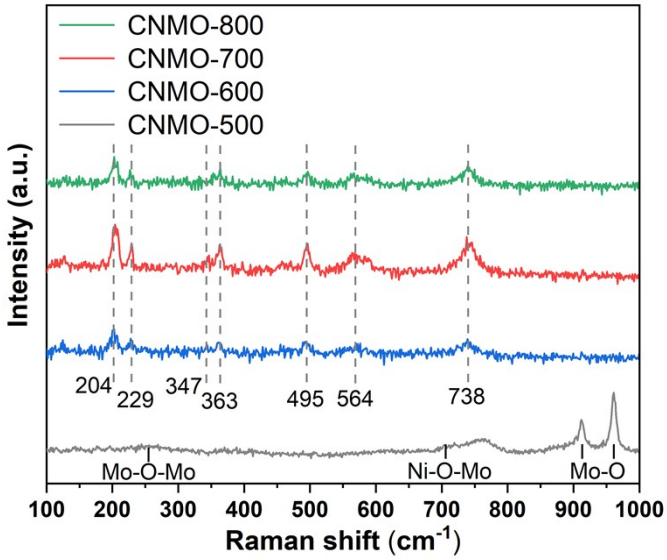

Fig. S4. Raman spectra of CNMOs at different calcination temperatures.

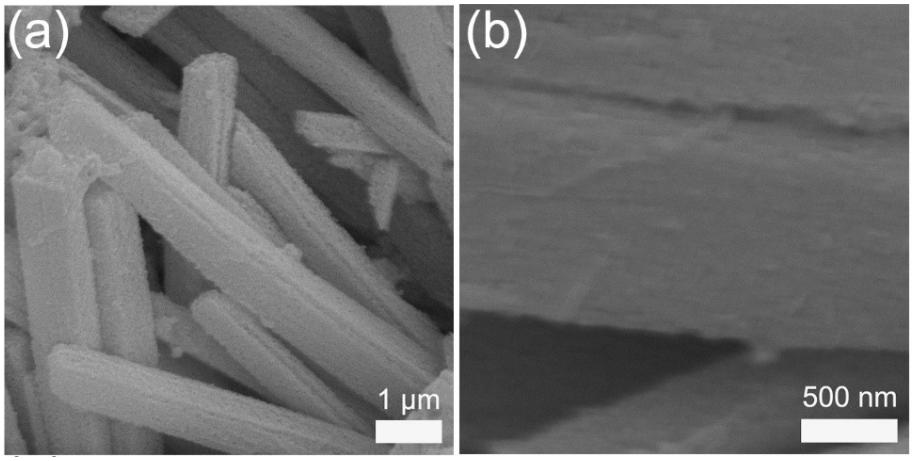

Fig. S5. SEM images of CNMO-700 after acid etching.

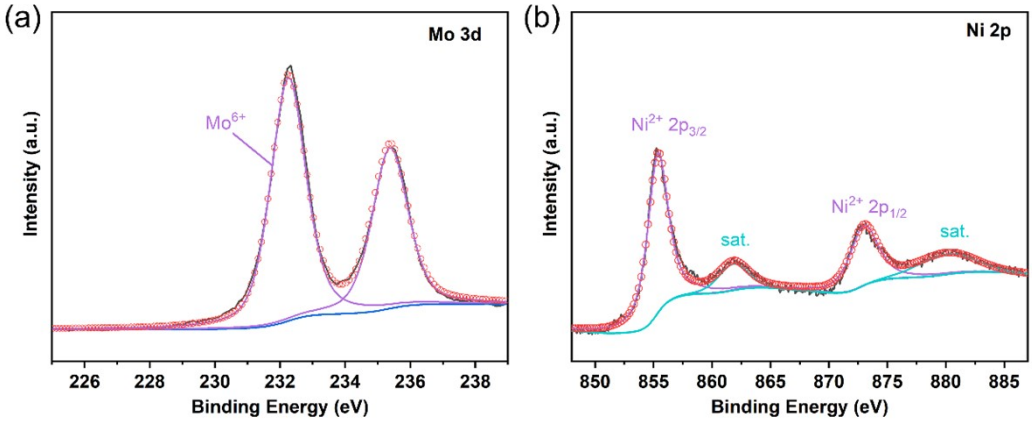

Fig. S6. XPS spectra of CNMO-500. (a) Mo 3d, (b) Ni 2p.

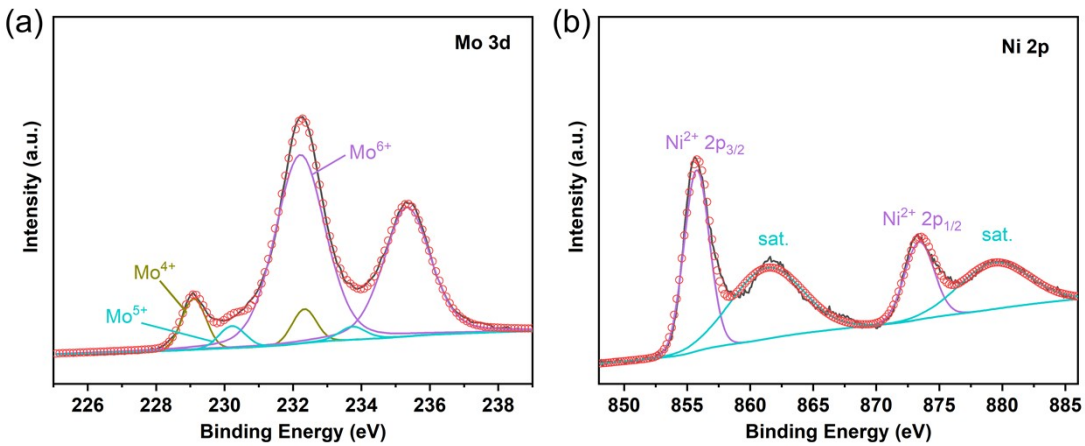

Fig. S7. XPS spectra of MoO_2. (a) Mo 3d, (b) Ni 2p.

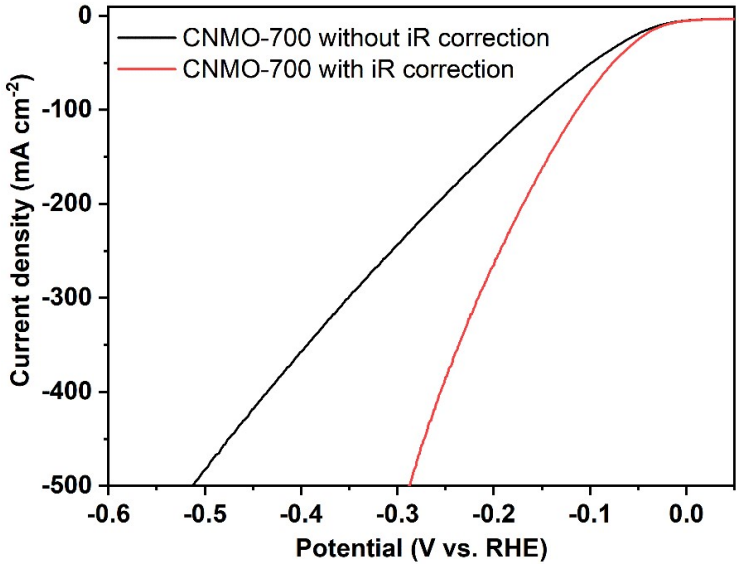

Fig. S8. LSV curves of CNMO-700 with/without iR correction for HER.

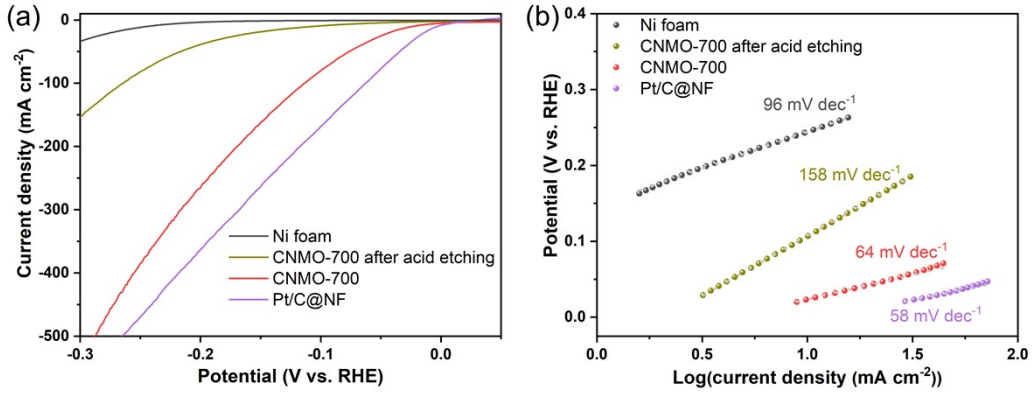

Fig. S9. (a) LSV curves and (b) Tafel slopes of Ni foam, CNMO-700 after acid etching, Pt/C and CNMO-700 for HER.

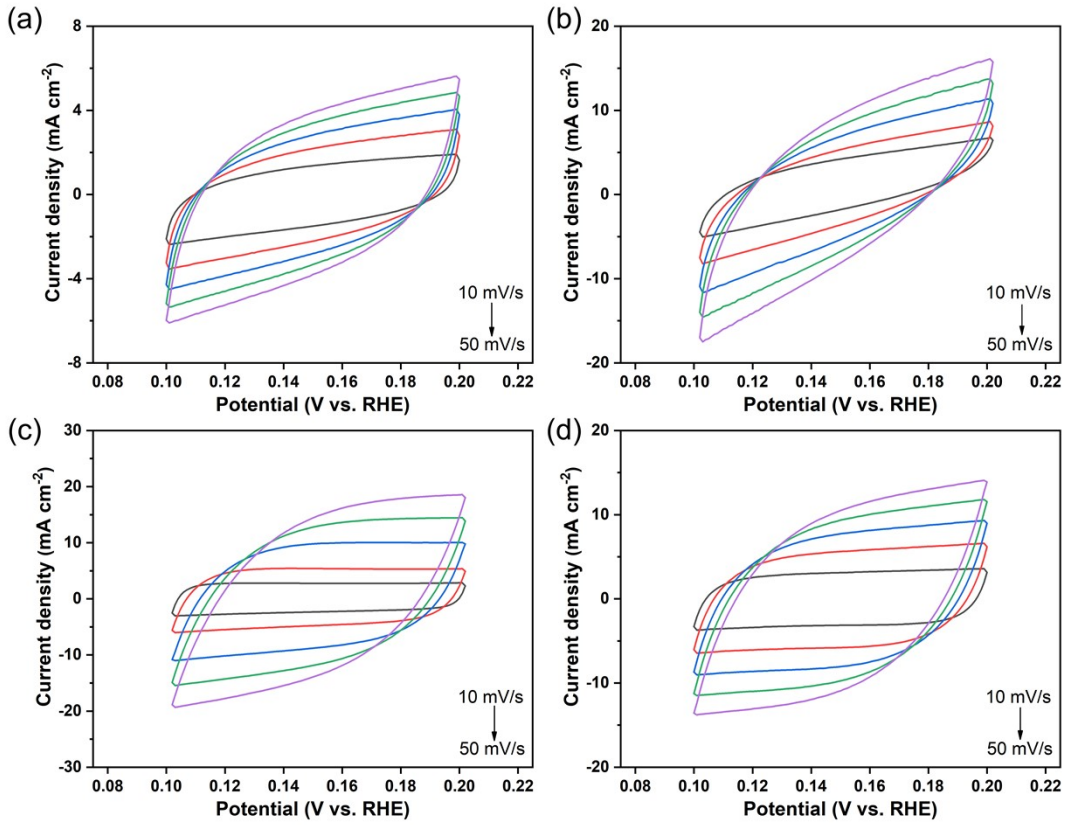

Fig. S10. Typical cyclic voltammograms of CNMO-500, 600, 700, 800 at scan rates ranging from 10 to 50 mV s^{-1}, the scanning potential range is from 0.10 V to 0.20 V.

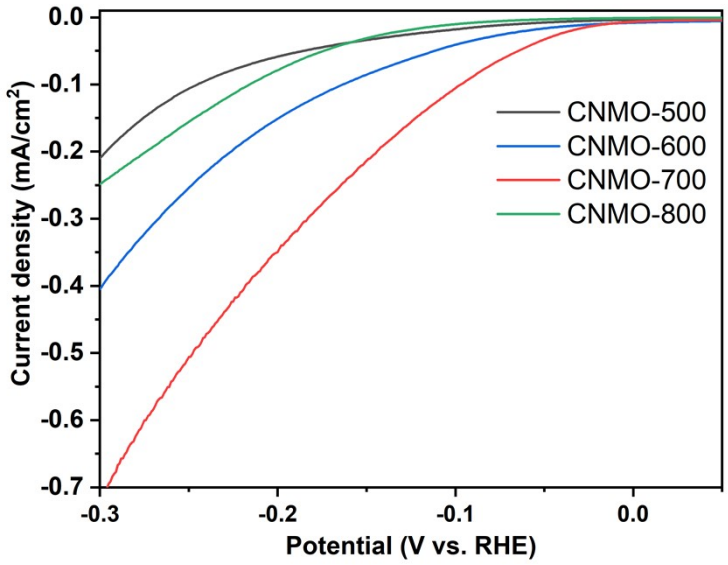

Fig. S11. HER activity (current density) of the samples normalized by their ECSAs.

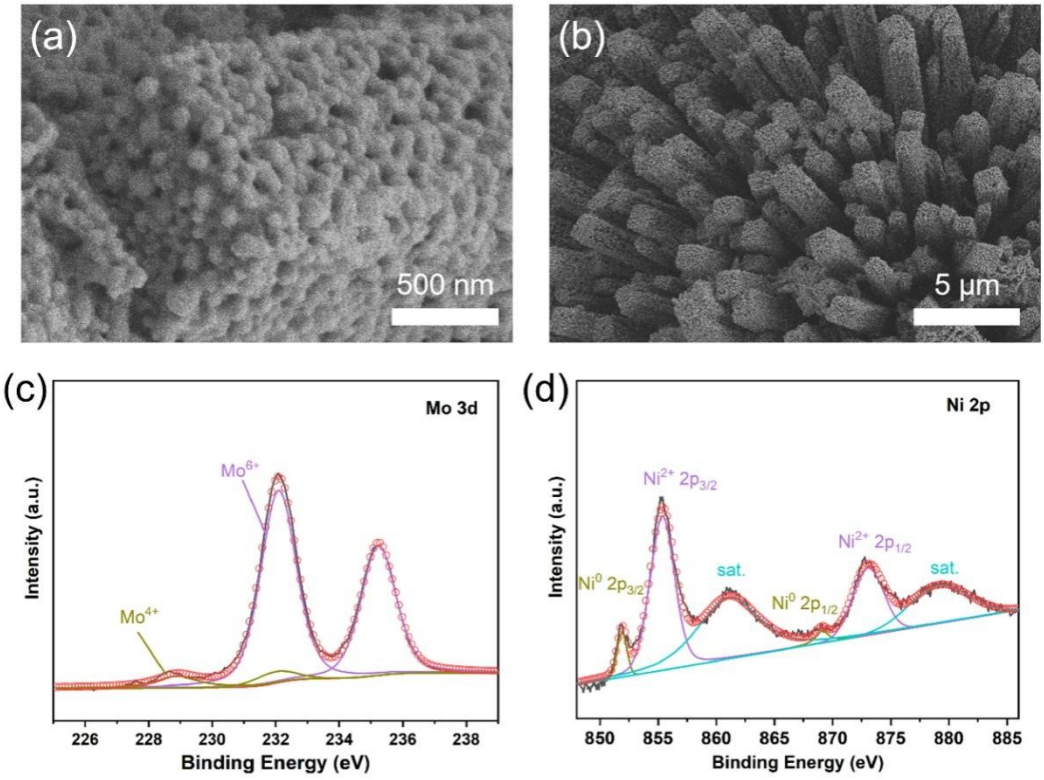

Fig. S12. (a, b) SEM images and (c, d) XPS spectra of CNMO-700 after hydrogen evolution stability test.

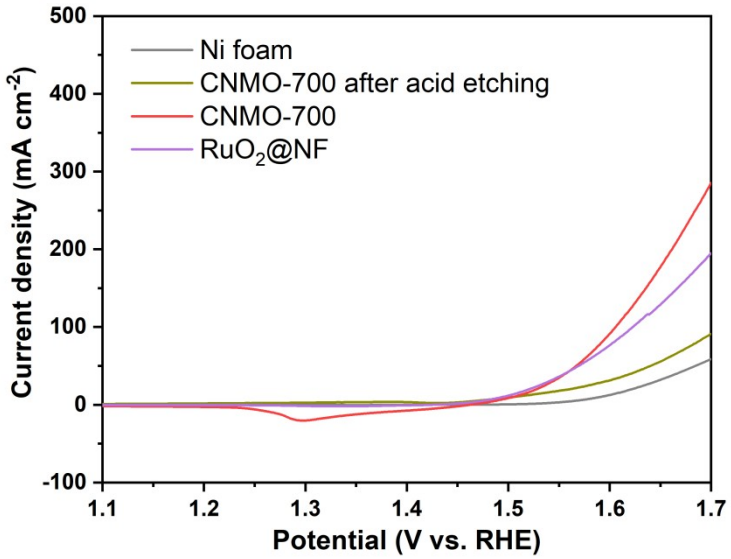

Fig. S13. LSV of Ni foam, CNMO-700 after acid etching, CNMO-700, RuO_2 for OER.

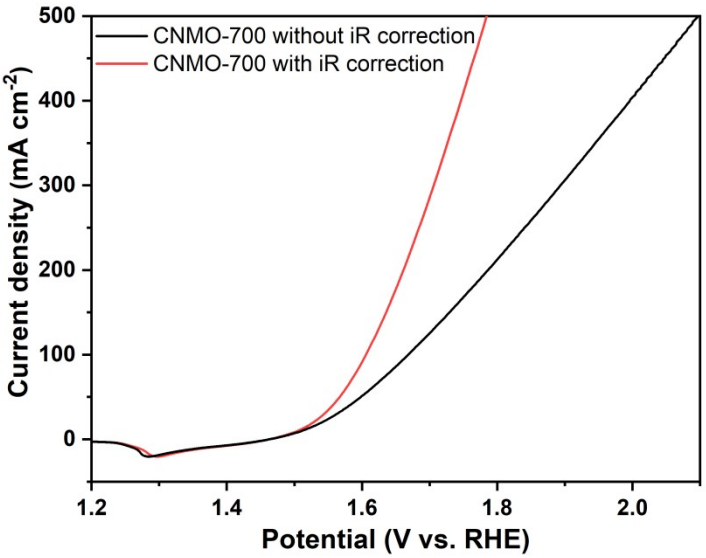

Fig. S14. LSV curves of CNMO-700 with and without iR correction for OER.

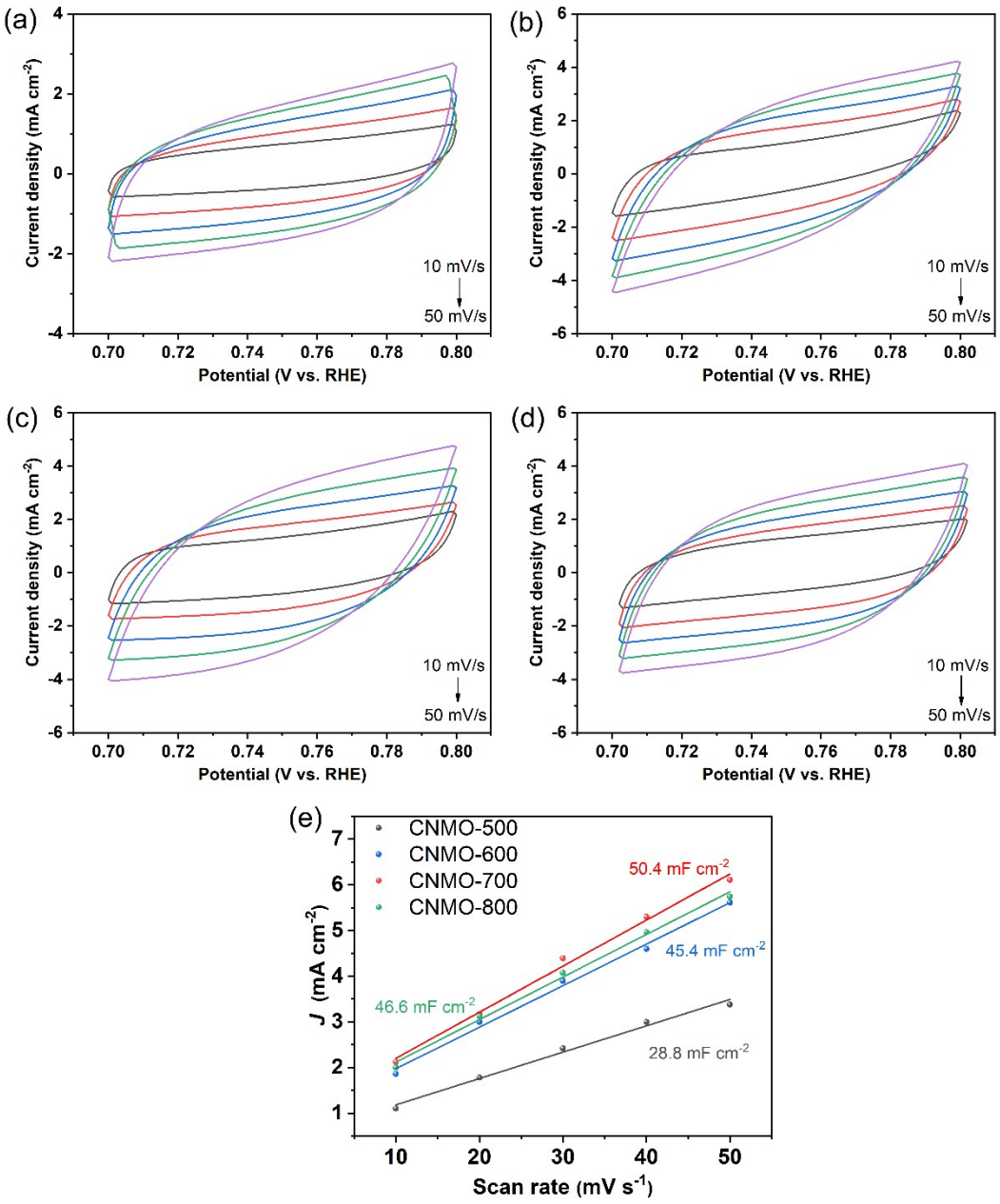

Fig. S15. (a-d) Typical cyclic voltammograms (CV) of CNMO-500, 600, 700, 800 at scan rates ranging from 10 to 50 mV s^{-1}, the scanning potential range is from 0.7 V to 0.8 V. (e) ECSA estimated by C_{dl} values.

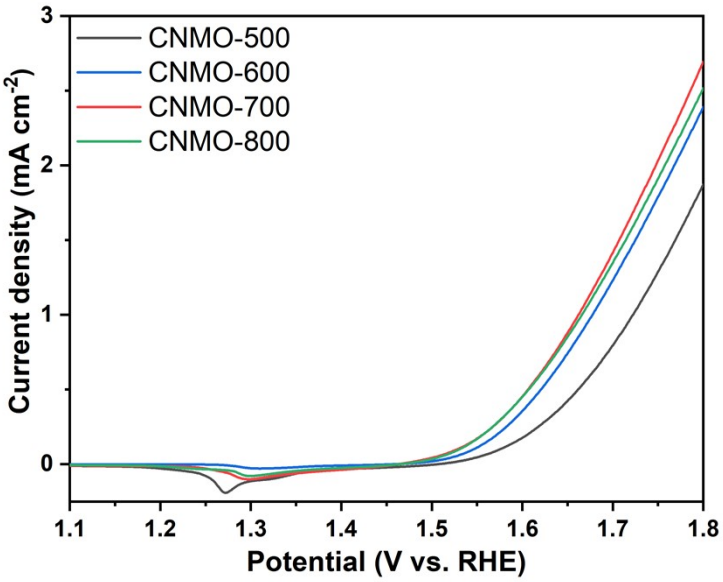

Fig. S16. OER activity (current density) of the samples normalized by their ECSAs.

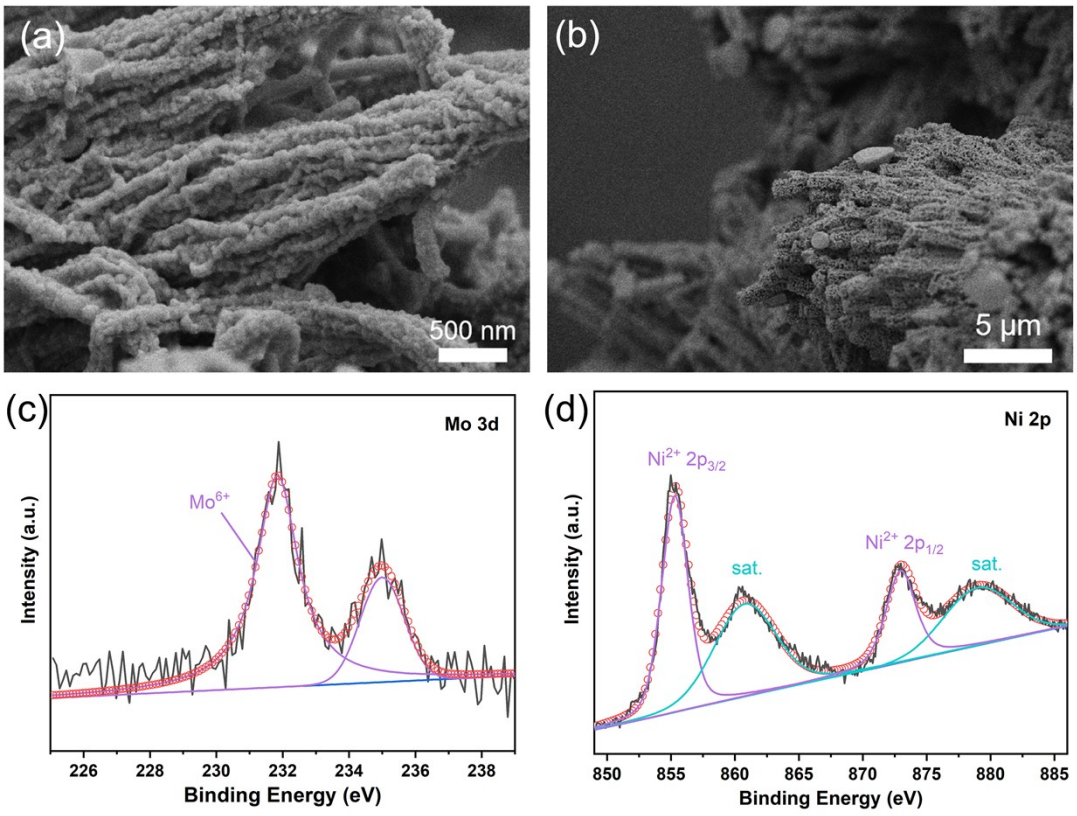

Fig. S17. (a, b) SEM images and (c, d) XPS spectra of CNMO-700 after OER stability test.

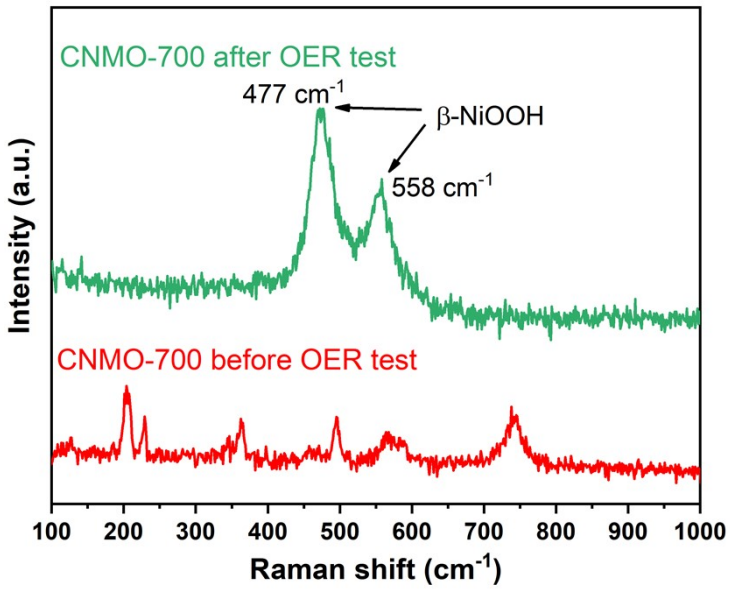

Fig. S18. Raman spectrum of the CNMO-700 before and after OER test.

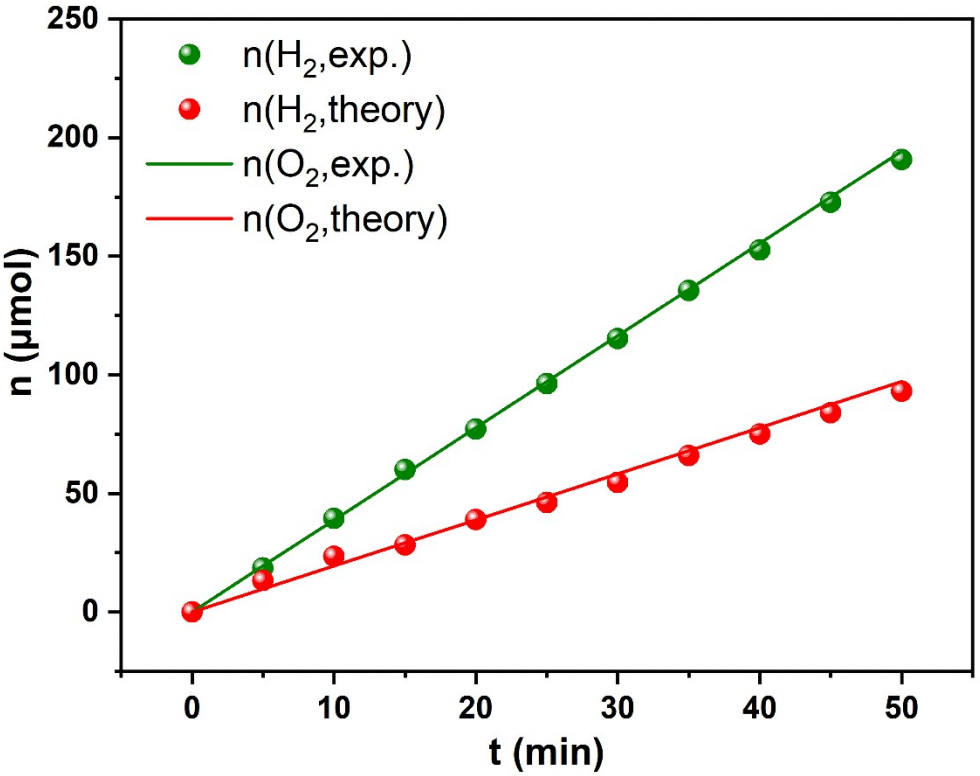

Fig. S19. The amount of gas theoretically calculated and experimentally measured versus time for overall water splitting at the current density of 50 mA cm^{-2}.

Table S1. Comparison of HER, OER and overall water splitting activity of Ni-MoO_2 with other TMO-based bifunctional electrocatalysts in 1 M KOH.

Materials	HER		OER		Overall	Ref.
	η_{10} (mV)	Tafel slope (mV dec^{-1})	η_{10} (mV)	Tafel slope (mV dec^{-1})	E_{10} (V)	
Ni-MoO_2	24	64	275	81	1.55	This work
NiO-Ni_3S_2	71	70	290	75	1.57	[1]
CF@Ni@NiO	153	84	300	60	1.65	[2]
NiFeP-MoO_2	56	81	149	29	1.41	[3]
Ni_2P/MoO_2@MoS_2	159	77	280	85	1.72	[4]
N-$NiMoO_4$/NiS_2	99	74	283	44	1.60	[5]
$NiMoO_{4-x}$/MoO_2	41	31	233	69	1.56	[6]
$NiCo_2O_4$	110	50	~280	53	1.65	[7]
Co-Fe NPs	220	73	340	51	1.92	[8]
$Ni_xCo_{1-x}MoO_4$@$CoMoO_4$	61	63	180	43	1.46	[9]
P-Co_3O_4	120	52	280	51.6	1.69	[10]
ceria/Ni-TMO	93	69	220	38	1.58	[11]
Co@Co_3O_4/FeNS-RGO	130	108	287	69	1.65	[12]
NF/H-$CoMoO_4$	42	91	295	/	1.56	[13]
CoNiMo-O	60	73	280	65	1.59	[14]

References.

1. L. Peng, J. Shen, X. Zheng, R. Xiang, M. Deng, Z. Mao, Z. Feng, L. Zhang, L. Li and Z. Wei, *Journal of Catalysis*, 2019, **369**, 345-351.
2. Z. Zhang, S. Liu, F. Xiao and S. Wang, *ACS Sustainable Chemistry & Engineering*, 2016, **5**, 529-536.
3. X. F. Wu, J. W. Li, Y. Li and Z. H. Wen, *Chemical Engineering Journal*, 2021, **409**.
4. Y. Wang, T. Williams, T. Gengenbach, B. Kong, D. Y. Zhao, H. T. Wang and C. Selomulya, *Nanoscale*, 2017, **9**, 17349-17356.
5. J. G. Li, K. Xie, H. Sun, Z. Li, X. Ao, Z. Chen, K. K. Ostrikov, C. Wang and W. Zhang, *ACS Appl Mater Interfaces*, 2019, **11**, 36649-36657.
6. Z. Zhang, X. Ma and J. Tang, *Journal of Materials Chemistry A*, 2018, **6**, 12361-12369.
7. X. Gao, H. Zhang, Q. Li, X. Yu, Z. Hong, X. Zhang, C. Liang and Z. Lin, *Angew Chem Int Ed Engl*, 2016, **55**, 6290-6294.
8. W. Adamson, X. Bo, Y. Li, B. H. R. Suryanto, X. Chen and C. Zhao, *Catalysis Today*, 2020, **351**, 44-49.
9. Z. K. Li, M. Y. Zheng, X. Zhao, J. Yang and W. L. Fan, *Nanoscale*, 2019, **11**, 22820-22831.
10. Z. Xiao, Y. Wang, Y.-C. Huang, Z. Wei, C.-L. Dong, J. Ma, S. Shen, Y. Li and S. Wang, *Energy & Environmental Science*, 2017, **10**, 2563-2569.
11. X. Long, H. Lin, D. Zhou, Y. An and S. Yang, *ACS Energy Letters*, 2018, **3**, 290-296.
12. J. Zhu, W. Tu, Z. Bai, H. Pan, P. Ji, H. Zhang, Z. Deng and H. Zhang, *Electrochimica Acta*, 2019, **323**.
13. K. Chi, X. Tian, Q. Wang, Z. Zhang, X. Zhang, Y. Zhang, F. Jing, Q. Lv, W. Yao, F. Xiao and S. Wang, *Journal of Catalysis*, 2020, **381**, 44-52.
14. B. Ren, D. Li, Q. Jin, H. Cui and C. Wang, *ChemElectroChem*, 2019, **6**, 413-420.